

Spin polarized transport through a single-molecule magnet: current-induced magnetic switching

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Magnetic switching of a single-molecule magnet (SMM) due to spin-polarized current is investigated theoretically. The charge transfer between the electrodes takes place *via* the lowest unoccupied molecular orbital (LUMO) of the SMM. Generally, the double occupancy of the LUMO level, and hence a finite on-site Coulomb repulsion, is allowed. Owing to the exchange interaction between electrons in the LUMO level and the SMM's spin, the latter can be reversed. The perturbation approach (Fermi golden rule) is applied to calculate current-voltage characteristics. The influence of Coulomb interactions on the switching process is also analyzed.

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I. INTRODUCTION

Single-molecule magnets (SMMs) are molecules with a relatively large net spin moment (corresponding to the spin number S) and a significant uniaxial magnetic anisotropy.^{1,2} As a result, behavior of SMMs resembles much that of superparamagnets, and at low temperatures the molecules become trapped in one of the two metastable spin states $|\pm S\rangle$.^{3,4,5,6} Owing to this bistability, SMMs seem to be a suitable base for memory cells of future information storage and processing technology.^{7,8} Apart from this, SMMs can possibly become basic components of the molecular-based spintronic devices.⁹

Electronic transport through an individual SMM has been demonstrated experimentally only very recently,^{11,12,13} attracting also a significant theoretical attention.^{14,15,16,17,18} An important issue in this context is the question of how the molecule's spin can be switched between the two stable states by means of spin-polarized current. This question is important not only from the purely fundamental reasons, but also from the point of view of possible applications of SMMs in various magnetoelectronic devices, and particularly as memory cells.

It is already well known that when a spin-polarized current flows through a magnet, some amount of spin momentum can be transferred from the current to magnetic body,¹⁹ leading effectively to a spin-transfer torque. This additional torque may lead to magnetic switching or current-induced precessional states. It has been shown recently, that exchange interaction between spin-polarized current and a SMM embedded in the barrier of a magnetic tunnel junction can lead to reversal of the molecule's spin.²⁰ The model considered there was simplified as the current was not flowing through the molecule, but rather directly between magnetic electrodes. However, the tunneling electrons could interact with the SMM *via* the exchange coupling, leading to switching of the SMM. The main objective of the present paper is to investigate theoretically a more realistic mechanism of SMM's switching, when the spin-polarized current flows directly through the molecule

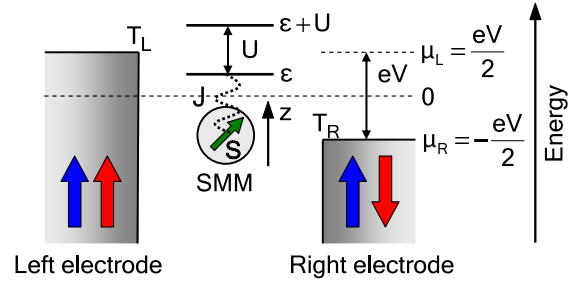


FIG. 1: (Color online) Schematic representation of the system under consideration in the nonequilibrium state, i.e. when a finite bias voltage V is applied, $eV = \mu_L - \mu_R$, where μ_L (μ_R) denotes the electrochemical potential of the left (right) electrode. Two collinear magnetic configurations of the leads' magnetic moments: parallel (black solid arrows) and antiparallel (grey solid arrows) are also indicated.

(molecular single-electron transistor geometry). In the model assumed, the current flows *via* the lowest unoccupied molecular orbital (LUMO) of the SMM. We restrict, however, our consideration to the case of the sequential transport regime. The results clearly show that transport of electrons through the LUMO level can lead to magnetic switching of the molecule, when the electrons in the LUMO level interact *via* exchange coupling with the spin moment of the inner core of the SMM.

In Sec. II we present the model and basis of the theoretical analysis of transport characteristics. Numerical results are presented in Sec. III. These results clearly show the possibility of magnetic switching induced by current pulse. Summary and final conclusions are in Sec. IV.

II. MODEL AND THEORETICAL DESCRIPTION

The model to be considered, see Fig. 1, consists of a SMM weakly coupled to two ferromagnetic electrodes (also called here leads). We assume that the electronic transport through the molecule takes place only *via* the

LUMO level of energy ε (measured from the Fermi level of the leads at equilibrium). This level is assumed to be exchange-coupled to the SMM's spin.⁸ Only collinear, i.e. parallel and antiparallel configurations of the leads' magnetic moments are considered, and these magnetic moments are assumed additionally to be parallel to the magnetic easy-axis of the molecule (along the axis z in Fig. 1).

The complete Hamiltonian of the system may be written as $\mathcal{H} = \mathcal{H}_{SMM} + \mathcal{H}_{el} + \mathcal{H}_T$. The first term describes the SMM and is assumed in the form,

$$\begin{aligned} \mathcal{H}_{SMM} = & - \left(D + \sum_{\sigma=\{\uparrow,\downarrow\}} \Delta D_1 c_{\sigma}^{\dagger} c_{\sigma} + \Delta D_2 c_{\uparrow}^{\dagger} c_{\downarrow}^{\dagger} c_{\downarrow} \right) S_z^2 \\ & + \sum_{\sigma=\{\uparrow,\downarrow\}} \varepsilon c_{\sigma}^{\dagger} c_{\sigma} + U c_{\uparrow}^{\dagger} c_{\uparrow} c_{\downarrow}^{\dagger} c_{\downarrow} \\ & - \frac{J}{2} \sum_{\sigma\sigma'=\{\uparrow,\downarrow\}} \boldsymbol{\sigma}_{\sigma\sigma'} \cdot \mathbf{S} c_{\sigma}^{\dagger} c_{\sigma'}, \end{aligned} \quad (1)$$

where \mathbf{S} is the SMM's spin operator, $\boldsymbol{\sigma}$ is the Pauli spin operator for electrons in the LUMO level, and $c_{\sigma}^{\dagger} (c_{\sigma})$ is the relevant electron creation (annihilation) operator. Apart from this, U denotes the Coulomb energy of two electrons of opposite spins in the LUMO level, whereas J is the exchange coupling parameter between the SMM's spin and electrons in the LUMO level. The parameter D is the uniaxial anisotropy constant of a free molecule, while ΔD_1 and ΔD_2 describe corrections to the anisotropy when the LUMO level is occupied by one and two electrons, respectively.²¹ The perpendicular anisotropy terms have been omitted as irrelevant for the effects described here. Apart from this, we neglect intrinsic spin relaxation, eg. that due to spin-phonon coupling. The only spin relaxation taken into account is that due to coupling of the molecule to the electrodes. This is justified as spin relaxation due to electronic processes associated with coupling of the dot to external leads is the dominant one.

The next term of \mathcal{H} describes the ferromagnetic electrodes,

$$\mathcal{H}_{el} = \sum_{q=\{L,R\}} \sum_{\mathbf{k},\sigma=\{\downarrow,\uparrow\}} \varepsilon_{\mathbf{k}\sigma}^q a_{\mathbf{k}\sigma}^{q\dagger} a_{\mathbf{k}\sigma}^q, \quad (2)$$

where $L(R)$ stands for the left (right) lead. The leads are characterized by non-interacting electrons with the dispersion relation $\varepsilon_{\mathbf{k}\sigma}^q$, where \mathbf{k} denotes a wave vector. In the equation above, $a_{\mathbf{k}\sigma}^q$ and $a_{\mathbf{k}\sigma}^{q\dagger}$ are the relevant annihilation and creation operators, respectively.

The final term of the Hamiltonian \mathcal{H} represents tunneling processes between the leads and the molecule,

$$\mathcal{H}_T = \sum_{q=\{L,R\}} \sum_{\mathbf{k},\sigma=\{\downarrow,\uparrow\}} \left[T_q a_{\mathbf{k}\sigma}^{q\dagger} c_{\sigma} + T_q^* c_{\sigma}^{\dagger} a_{\mathbf{k}\sigma}^q \right], \quad (3)$$

where T_L and T_R are the tunneling matrix elements between the SMM and the left and right electrodes, respectively. These parameters are assumed to be independent

of the wave vector and spin orientation. We point, that direct electron tunneling between the leads is excluded.

It has been shown⁸ that the Hamiltonian \mathcal{H}_{SMM} , Eq. (1), commutes with the z component S_t^z of the total spin operator $\mathbf{S}_t \equiv \mathbf{S} + \sum_{\sigma\sigma'} \boldsymbol{\sigma}_{\sigma\sigma'} c_{\sigma}^{\dagger} c_{\sigma'}/2$, where the second term of \mathbf{S}_t represents the spin of an electron in the LUMO. As a consequence, if one treats \mathcal{H}_{SMM} as the unperturbed part of the total Hamiltonian \mathcal{H} , it is convenient to numerate the eigenstates of \mathcal{H}_{SMM} with the eigenvalues m of S_t^z and with the number of electrons in the LUMO level. Thus, the eigenstates of the SMM are given by: $|0, m\rangle \equiv |0\rangle_o \otimes |m\rangle_{mol}$, $|2, m\rangle \equiv |\uparrow\downarrow\rangle_o \otimes |m\rangle_{mol}$, $|1, m\rangle^{\pm} \equiv \mathbb{A}_m^{\pm} |\downarrow\rangle_o \otimes |m+1/2\rangle_{mol} + \mathbb{B}_m^{\pm} |\uparrow\rangle_o \otimes |m-1/2\rangle_{mol}$ for the intermediate states, and $|1, \pm S \pm 1/2\rangle \equiv |\uparrow (\downarrow)\rangle_o \otimes |\pm S\rangle_{mol}$ for the fully polarized states. According to our notation, $|\bullet\rangle_{o(mol)}$ denotes the spin state of the orbital (SMM). The coefficients \mathbb{A}_m^{\pm} and \mathbb{B}_m^{\pm} act here as effective Clebsch-Gordan coefficients which depend on the system's parameters, and have the form

$$\mathbb{A}_m^{\pm} = \mp \frac{\sqrt{2\Delta\epsilon(m) \pm (2D^{(1)} - J)m}}{2\sqrt{\Delta\epsilon(m)}}, \quad (4)$$

$$\mathbb{B}_m^{\pm} = \frac{J\sqrt{S(S+1) - m^2 + 1/4}}{2\sqrt{\Delta\epsilon(m)}\sqrt{2\Delta\epsilon(m) \pm (2D^{(1)} - J)m}}, \quad (5)$$

where $\Delta\epsilon(m) = \sqrt{D^{(1)}(D^{(1)} - J)m^2 + (J/4)^2(2S+1)^2}$ and $D^{(1)} \equiv D + \Delta D_1$. Additionally, we assume $2D^{(1)} - J \geq 0$. The corresponding eigenenergies of the Hamiltonian \mathcal{H}_{SMM} are: $\epsilon(0, m) = -Dm^2$, $\epsilon(2, m) = 2\varepsilon + U - (D + 2\Delta D_1 + \Delta D_2)m^2$ and $\epsilon(1, m)^{\pm} = \varepsilon + J/4 - (D + \Delta D_1)(m^2 + 1/4) \pm \Delta\epsilon(m)$. The energy of the fully polarized states $|1, \pm S \pm 1/2\rangle$ is $\epsilon(1, \pm S \pm 1/2)^{\pm}$.

To investigate the current-induced magnetic switching of the SMM, we analyze the relevant I-V characteristics. The total current flowing through the molecule can be written as $I = (I_L - I_R)/2$, where I_{α} ($\alpha = L, R$) denotes the current flowing from the lead α to the molecule,

$$I_{\alpha} = e \sum_{m_r, m_q} \sum_{n_r, n_q} (n_r - n_q) \gamma_{\alpha}^{|n_q, m_q\rangle |n_r, m_r\rangle} P_{|n_q, m_q\rangle}. \quad (6)$$

Here, $\gamma_{\alpha}^{|n_q, m_q\rangle |n_r, m_r\rangle}$ represents the rate of transitions between the states $|n_q, m_q\rangle$ and $|n_r, m_r\rangle$, whereas $P_{|n_q, m_q\rangle}$ is the probability of finding the SMM in the state $|n_q, m_q\rangle$. We assume that current is positive when electrons flow from the left to right. For notational clarity, from now on we assume $|n_q, m_q\rangle \equiv |q\rangle$, which also means that $\sum_q \equiv \sum_{n_q} \sum_{m_q}$.

To find current we need to determine first both the transition rates $\gamma_{\alpha}^{|q\rangle |r\rangle}$ and the probabilities $P_{|q\rangle}$. Let us start with the transition rates. In the second order (Fermi golden rule) one finds $\gamma_{\alpha}^{|q\rangle |r\rangle}$ in the form,

$$\gamma_{\alpha}^{|q\rangle |r\rangle} = \sum_{k, \sigma \in \alpha} \left[W_{|r\rangle}^{k\sigma|q} f(\varepsilon_{k\sigma}^{\alpha}) + W_{|q\rangle}^{k\sigma|r} [1 - f(\varepsilon_{k\sigma}^{\alpha})] \right], \quad (7)$$

where the first term corresponds to electron transitions from the α -th lead to the molecule, while the second term

describes the charge transfer back to the lead α . Furthermore, $f(\varepsilon)$ is the Fermi-Dirac distribution function, and $W_f^i = (2\pi/\hbar)|\langle f|\mathcal{H}_T|i\rangle|^2\delta(E_f - E_i)$ is the rate of transitions from an initial state (i) to a final state (f).

The final expression for the transition rates $\gamma_{\alpha}^{[q]|r}$ takes the form

$$\gamma_{\alpha}^{[q]|r} = \frac{1}{\hbar} \sum_{\sigma=\{\downarrow,\uparrow\}} \Gamma_{\sigma}^{\alpha} \left\{ |C_{qr}^{\sigma}|^2 f(\epsilon(r) - \epsilon(q) - \mu_{\alpha}) + |C_{rq}^{\sigma}|^2 \left[1 - f(\epsilon(q) - \epsilon(r) - \mu_{\alpha}) \right] \right\}, \quad (8)$$

where $\Gamma_{\sigma}^{\alpha} = 2\pi|T_{\alpha}|^2 D_{\sigma}^{\alpha}$ is the LUMO level width acquired due to coupling of the level to the lead α , and D_{σ}^{α} denotes the spin-dependent density of states (DOS) at the Fermi level in the α -th electrode. These parameters will be used in the following to describe strength of the coupling between the SMM and leads. It is convenient to write Γ_{σ}^{α} as $\Gamma_{\pm}^{\alpha} = \Gamma_{\alpha}(1 \pm P_{\alpha})$, where $\Gamma_{\alpha} = (\Gamma_{+}^{\alpha} + \Gamma_{-}^{\alpha})/2$, and P_{α} is the spin polarization of the lead α , $P_{\alpha} = (D_{+}^{\alpha} - D_{-}^{\alpha})/(D_{+}^{\alpha} + D_{-}^{\alpha})$. Here $\sigma = +(-)$ corresponds to spin-majority (spin-minority) electrons. In the following we assume that the couplings are symmetric, $\Gamma_L = \Gamma_R = \Gamma/2$. Finally, in Eq. (8) $|C_{qr}^{\sigma}|^2 \equiv |\langle r|c_{\sigma}^{\dagger}|q\rangle|^2$ together with $|C_{rq}^{\sigma}|^2 \equiv |\langle r|c_{\sigma}|q\rangle|^2$ constitute basic selection rules for transitions between neighboring molecular states. The transition is allowed only when the charge state of the SMM is changed by one and the change in the total spin satisfies $\Delta S_t^z = \pm 1/2$. Assuming that the SMM is initially saturated in the state $|0, -S\rangle$, see Fig. 2, one may expect that at a sufficiently large voltage the molecule can be switched to the final state $|0, S\rangle$. The switching process corresponds then to the reversal of the SMM's spin *via* all the intermediate states.

The probabilities $P_{|q\rangle}$ (see Eq. (6)), are obtained from the master equations

$$c \frac{dP_{|q\rangle}}{dV} = \sum_{\alpha} \sum_r \left[\gamma_{\alpha}^{[r]|q} P_{|r\rangle} - \gamma_{\alpha}^{[q]|r} P_{|q\rangle} \right], \quad (9)$$

for $n_q = 0, 1^{(\pm)}, 2$ and $m_q \in \langle -S-1/2, S+1/2 \rangle$ (we recall the definition, $|n_q, m_q\rangle \equiv |q\rangle$). In the present paper, we assume that the voltage is augmented linearly in time, $V = ct$, with c denoting the speed at which the voltage is increased. The corresponding time scale, however, is much slower than that set by electronic transitions. The relevant boundary conditions for the probabilities $P_{|q\rangle}$ are: $P_{|0, -S\rangle}(V = 0) = 1$ and $P_{|q\rangle}(V = 0) = 0$ for $|q\rangle \neq |1, -S\rangle$.

III. NUMERICAL RESULTS AND DISCUSSION

The results have been computed for an octanuclear iron(III) oxo-hydroxo cluster of the formula $[\text{Fe}_8\text{O}_2(\text{OH})_{12}(\text{tacn})_6]^{8+}$ (shortly Fe_8). Such a free-standing molecule has spin corresponding to $S = 10$. The following values of the molecule parameters are taken:

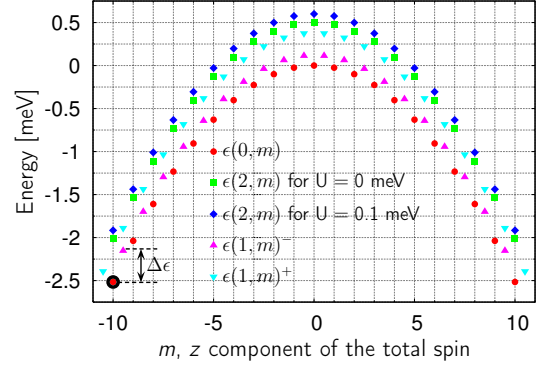


FIG. 2: (Color online) The energy levels of a Fe_8 molecular magnet for two values of the Coulomb parameter U . The black bold circle indicates the initial state $|0, -S\rangle$, and $\Delta\epsilon = 0.36$ meV is the energy gap corresponding to the activation energy for the magnetic switching.

$D = 0.292$ K ($D \approx 0.025$ meV),²² $J = 0.025$ meV and $\epsilon = 0.25$ meV. Since there is no clear and reliable experimental evidence of the change in anisotropy constant of the Fe_8 molecule due to extra electrons in the LUMO level, we assume $\Delta D_1 = \Delta D_2 = 0$. The couplings of the molecule to the left and right lead are assumed to be the same, $\Gamma_L = \Gamma_R = 0.0015$ meV. We also assume that both the electrodes are made of the metallic material characterized by the same polarization parameter P , $P = P_L = P_R$. The calculations have been performed for the temperature $T = 0.01$ K, which is below the blocking temperature $T_B = 0.36$ K. The corresponding energy levels of the molecule are shown in Fig. 2. It is worth noting that for the parameters assumed, the ground spin state of the molecule attached to the leads ($S_t^z = \pm 10$) is the same as that of a free-standing molecule ($S_z = \pm 10$). Nevertheless, for a sufficiently low energy of the LUMO level, which can be controlled for instance with a gate voltage, the ground state of the molecule attached to the leads can correspond to $S_t^z = \pm 21/2$ (the molecule with one extra electron on the LUMO level).

Fig. 3 presents the average $\langle S_t^z \rangle$ and current I flowing through the system for different values of the Coulomb parameter U in both parallel and antiparallel magnetic configurations of the leads. It can be noted that the reversal of the SMM's spin occurs only in the antiparallel configuration, whereas in the parallel configuration all molecular spin states gradually become equally probable. As a consequence, $\langle S_t^z \rangle \rightarrow 0$ and the magnetic switching is not observed. This is a consequence of the left/right symmetry of the molecule's coupling to external leads – similarly to the absence of spin accumulation in tunneling through a metallic nanoparticle in the parallel magnetic configuration. However, such a symmetry is absent in the antiparallel configuration, and accordingly the spin states of the molecule become unequally occupied, which in turn results in spin reversal.

The corresponding current-voltage characteristics, shown in Fig. 3 by the dashed lines, reveal features (steps

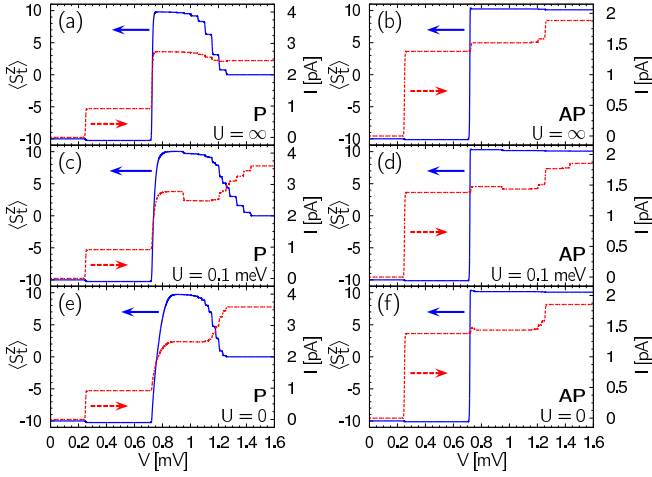


FIG. 3: (Color online) The average value of the total spin $\langle S_z^T \rangle$ (solid line) and the current I flowing through the system (dashed line) in the case of parallel (P) and antiparallel (AP) magnetic configurations for different values of the Coulomb parameter U . The other parameters are: $P_L = P_R = 0.5$ and $c = 1$ V/s.

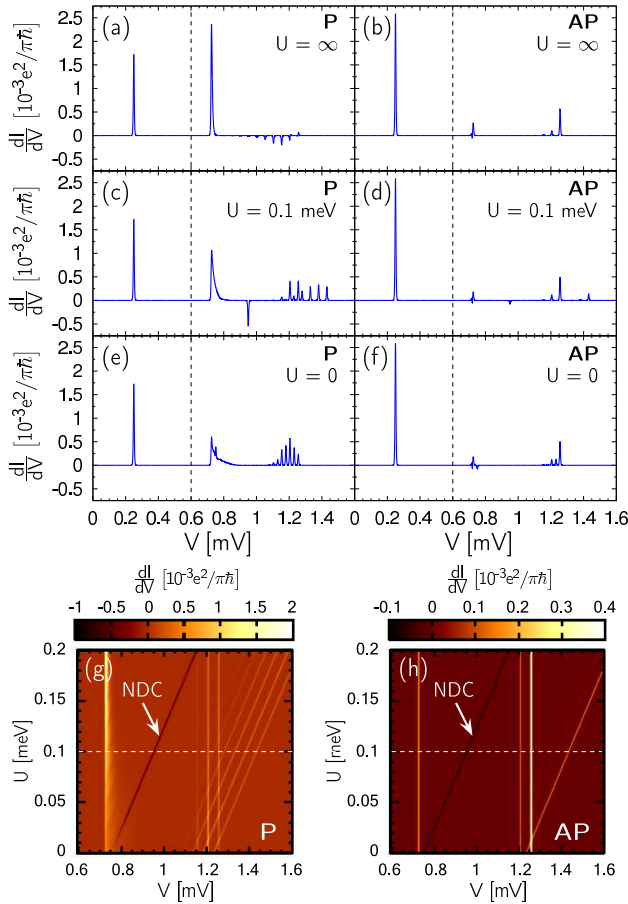


FIG. 4: (Color online) Differential conductance dI/dV for two collinear, i.e. parallel (P) and antiparallel (AP) magnetic configurations. The parameters as in Fig. 3.

in the current) which are directly related to the reversal process. In fact, each step corresponds to a certain type of transitions between neighboring molecular spin levels. Consider for instance the main features of the I - V plots shown in Fig. 3. The plots are almost the same as long as $V < 0.72$ mV, i.e. for eV below the energy activating the reversal process. The first step corresponds to the transition between the states $|0, -10\rangle$ and $|1, -21/2\rangle$ (see also Fig. 2). As voltage increases further, the next step appears due to transitions between the states $|0, -10\rangle$ and $|1, -19/2\rangle^-$, and the magnetic switching begins. Augmenting voltage further, one finds certain regions of bias voltage, where current drops with increasing bias. This behavior is particularly visible in the parallel configuration, see Figs. 3 (a,c). The drop in current with increasing bias is equivalent to negative differential conductance (NDC), see also Fig. 4. The negative differential conductance is a consequence of the spin blockade phenomenon²³, which in turn follows from the inequality of the transition rates to the two levels. The suppression of the current starts when the system becomes energetically allowed to transfer from the state $|1, -21/2\rangle$ to the state $|2, -10\rangle$.

The differential conductance corresponding to the I - V curves shown in Fig. 3 is presented in Fig. 4. The peaks correspond to the steps from Fig. 3. The negative differential conductance due to spin blockade is clearly seen, particularly in the parallel configuration for $U > 0$, although some weak negative peaks also occur in the antiparallel case. Evolution of the differential conductance with the Coulomb energy U and the bias voltage V is shown explicitly in Figs. 4 (g)-(h) for both magnetic configurations of the leads.

IV. SUMMARY

In this paper we have considered electronic transport through a single molecular magnet attached to ferromagnetic leads. The molecule is characterized by a spin number S and an additional unoccupied orbital, which becomes active in transport through the molecule.

We have shown that spin polarized electrons tunneling through the LUMO level of a SMM can reverse the SMM's spin when the electrons in the LUMO level interact *via* exchange coupling with the SMM's spin. The reversal starts at a certain threshold voltage corresponding to the distance between the two lowest energy levels. It is interesting to note, that for symmetrical systems, the spin reversal takes place only in the antiparallel configuration. The conductance spectra also show regions of negative differential conductance due to spin blockade effect.

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